

Spontaneous growth of an InAs nanowire lattice in an InAs/GaSb superlattice

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Abstract

We describe a lattice of InAs nanowires that spontaneously organizes in three dimensions within an InAs/GaSb superlattice grown under high As₄ flux. As characterized by x-ray diffraction and cross-sectional scanning tunneling microscopy, the periodic nanowires are ~10 nm high, 120 nm wide, and many microns long along [110], with face-centered cubic-like vertical ordering within the superlattice. The unusual vertical ordering creates a lateral composition modulation with half the period of the nanowires. The structure appears to arise from the InAs misfit stress combined with specific InAs and GaSb growth kinetic effects.

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The growth of a strained-layer superlattice is inherently unstable, leading to a variety of possible modulations in the film morphology.^{1,2} Depending on the number of components in a heterostructure and the growth conditions, lateral modulations may be exhibited in the film thickness or elemental composition. Moreover, the long-range strain fields associated with these modulations may exert an influence on subsequent growth, vertically correlating the lateral modulations within the superlattice; these correlations are well explained by linear stability theory.^{1,2} Although growth instabilities are often detrimental to the desired superlattice material properties, it is now widely recognized that they can be used to create potentially useful low-dimensional nanostructures—e.g. quantum dots and wires—that are ordered in three dimensions. For Si-Ge and III-V strained-layer superlattices, quantum dots and wires caused by growth instabilities typically align vertically one-atop-the-other.¹ Here we describe the spontaneous organization of a periodic lattice of microns-long InAs nanowires within an InAs/GaSb superlattice, where the vertical ordering has an unusual face-centered cubic-like structure, i.e. the wires are vertically anti-correlated.

InAs/GaSb superlattices (SLs) were grown by molecular beam epitaxy (MBE) on GaSb substrates at ~390 °C using Sb₄ and either As₂ or As₄ from a valved arsenic cracker. Except where otherwise noted, each SL was comprised of >100 periods of 13 monolayers (ML) of InAs alternating with 13 ML of GaSb, with InSb bonds at each interface. The growth rate for both materials was 0.5 ML/s. The samples were characterized by double-crystal x-ray diffraction (XRD) and cross-sectional scanning tunneling microscopy (XSTM). For XSTM, each sample was scribed and cleaved in the ultra-high vacuum chamber immediately prior to characterization. All STM images shown are of filled electronic states recorded with constant currents of 0.1–

0.2 nA. Additional details of the growth and sample preparation procedures have been reported elsewhere.^{3,4}

In a previous study of the effects of As_4 versus As_2 on InAs/GaSb heterostructures, we reported that the use of high As_4 fluxes can lead to unusual morphological modulations in the SL.⁴ For SLs grown with an As_4 :In flux ratio greater than 2:1, modulations were evident in the occurrence of “extra” satellite peaks in the XRD spectra and film thickness variations apparent from XSTM (modulations never observed with As_2). As observed in the XRD spectrum in Fig. 1(a), satellites occur midway between the principal SL diffraction peaks, indicating a doubling of the expected 8 nm period. The specific nature of the period doubling can be seen in STM images of the (110) cross-sectional plane through the SL (Fig. 1). Fig. 1(b) reveals the nanoscale structure starting from the GaSb buffer layer through the first 80 SL periods. Under these imaging conditions, the GaSb layers appear as bright bands and the InAs as dark bands. It is apparent that the growth became unstable almost immediately, because a lateral modulation in the InAs layer thickness is already evident in the second and third InAs periods. A well-ordered modulation is in place by about the 20th SL period, with a lateral period of 120 nm in each layer, and a vertical period of 16 nm (exactly twice the SL period).

The doubling of the SL period is caused by an unusual vertical alignment of the thickness modulations. Typically, lateral thickness modulations in SLs composed of materials A and B develop in one of two ways.² In the “in-phase” mode ($\varphi = 0^\circ$), B’s modulation occurs directly on top of A’s, leading to progressively rougher (and unstable) growth. In the “out-of-phase” mode ($\varphi = 180^\circ$), layer B is thickest where A is thinnest (and vice-versa), creating a periodic, lateral segregation of the two materials that maintains the SL period. In our case, each InAs layer has a strong lateral thickness modulation, nearly vanishing at its thinnest point. The GaSb layers have

a much smaller thickness modulation, with the layer thinner at the top of the InAs modulation, approximately the intended thickness at the bottom of the InAs modulation, but thicker *on the sides* of the InAs modulation ($\varphi = 90^\circ$). Each GaSb layer has the effect of slightly flattening the amplitude of the underlying InAs morphological modulation while maintaining its lateral phase. However, the next InAs layer shifts 180° from the previous InAs layer, growing thickest where the previous InAs layer is *thinnest*. As viewed across the (110) plane, the net effect is a face-center cubic (fcc)-like structure with a vertical period twice that of the SL, in the sequence thick InAs—thin GaSb—thin InAs—normal GaSb—thick InAs.

The atomic scale structure of the composition modulation can be seen in Fig. 1(c). Under these imaging conditions, XSTM essentially shows a cross-section through one As and Sb lattice plane of every-other InAs or GaSb monolayer grown; i.e. every 26 ML SL period is observed as 13 rows in the image. A number of observations can be made. First, with the exception of some Sb cross-incorporated into the InAs (intrinsic to this material system^{3,5}), there is no wide-scale III-V segregation or alloying between or within the layers. It is also clear that the InAs modulation reduces the InAs thickness to as little as 1–2 ML in the troughs. Although the modulation is not well ordered in this particular region, the area outlined highlights thickening of the GaSb along the stepped edge of the InAs mound that is characteristic of a vertically well-ordered structure.

The three-dimensional structure of the SL was determined by examining three orthogonal lattice planes (Fig. 2). The structural modulation is highly anisotropic, consisting of InAs nanowires oriented along the [110] crystallographic direction, as illustrated in Fig. 2(b). This structure is most clear in the topography of the growth surface, as shown in the atomic force microscopy image in Fig. 2(a), where 120 nm-wide “wires” are observed to extend for many

microns of length. Because of the wire-like structure, in XSTM images of the $(\bar{1}10)$ face the modulation is only evident as a vertically periodic variation of the films' thicknesses [Fig. 2(c)]. The unusual fcc-like vertical alignment creates an overall lateral composition modulation with a period *half* that of the modulation within each layer. The net composition modulation is very apparent in the prominent artifact it causes in XSTM images of the (110) surfaces: a 60 nm-period wave across the surface where the InAs- and GaSb-rich regions structurally relax inward and outward from the surface, respectively, in order to accommodate the local strain.⁶ To our knowledge, only one other III-V SL structure has been described that even resembles this one, an InAs/InAlAs SL on InP(001),⁷ where shorter “wires” oriented perpendicular to those we observe occur with less consistent vertical ordering.

Because misfit stress is often the cause of growth instability in heteroepitaxy, variations of the original InAs/GaSb SLs characterized above were grown with the same As₄ flux but different periods or substrates, with interesting results (Fig. 3). One SL was grown with GaSb layers 26 ML thick instead of 13 ML. As shown in Fig. 3(a), a qualitatively similar vertically ordered composition modulation is observed, but with a *larger* lateral period, ~150 nm. Another sample was grown on a relaxed AlSb buffer layer on the GaSb substrate, increasing the effective substrate lattice constant by 0.66%. The same composition modulation structure is again observed [Fig. 3(b)], with a relatively *smaller* lateral period, ~100 nm. The final sample examined, shown in Fig. 3(c), was grown on an InAs buffer and substrate, which decreases the lattice constant by 0.61% compared with GaSb. In addition, the InAs and GaSb layers were coupled with GaAs interfacial bonds instead of InSb, reducing the average vertical lattice constant in the SL. Under these strain conditions, the growth instability caused by high As₄ pressure is not observed, just the expected SL heterostructure.

The unusual structure of the InAs/GaSb SLs results from a complex convergence of strain-related thermodynamic and growth kinetic effects. Although a definitive explanation is beyond the scope of this report, it is possible to make qualitative sense of many of the results. Thin InAs films grown very slowly on GaSb exhibit a morphological roughness with a wavelength, $\lambda = 140$ nm, close to what we observe for the SL, and consistent with linear stability theory (which predicts λ decreases with increasing strain).⁸ Growing on the larger lattice of an AlSb buffer increases the misfit of the InAs layers within the SL, decreasing λ ; an InAs substrate essentially eliminates the misfit and therefore the related instability. The change with GaSb layer thickness can also be reconciled if one considers both the vertical and lateral strains. Although the SL is laterally coherent with the GaSb substrate, the InSb interfacial bonds and Sb incorporated into the InAs make the average vertical lattice constant of the SL *larger* than the substrate. Doubling the thickness of each GaSb layer dilutes this increase. If the comparatively smaller vertical lattice reduces the overall tensile strain in the InAs, it would account for the larger λ .

The occurrence of the instability under relatively high As₄ flux, but not As₂, arises from both strain and kinetic effects. Growing with As₄ suppresses the cross-incorporation of As into GaSb that occurs with As₂.⁴ With less As in the GaSb, the average vertical lattice constant in the SL is larger, increasing the overall InAs strain and promoting the instability. Just as important, high As pressure appears to increase the In diffusion length and promote step-flow growth,⁹ making it easier for the thermodynamically-favored morphology to be reached. Assuming this morphology is a near-equilibrium structure, the wire-like shape must arise from the inherent structural anisotropy of the (001) III-V surface. There is theoretical evidence that InAs steps parallel to the [110] direction preferentially reduce the strain energy,¹⁰ possibly accounting for the shape.

The unusual vertical alignment of the lateral morphological oscillations arises from the 90° phase shift of alternate layers in the SL. The GaSb shift on the InAs occurs because the GaSb preferentially grows on the {n11} vicinal sides of the InAs wires. Although little is known about the growth of GaSb on faceted surfaces, there is at least one report that growth is, indeed faster on such facets than on the (001) surface (albeit for metalorganic MBE).¹¹ The 90° GaSb shift forces the subsequent InAs modulation to shift 90° as well in order to maintain the energetically favored volume segregation of the two SL components. If this were not the case, the lateral modulation amplitude would grow and the SL growth would become unstable. Therefore, it is possible that the kinetics of GaSb growth on vicinal surfaces is the key component in the organization of this most unusual self-organized nanostructure.

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Figures

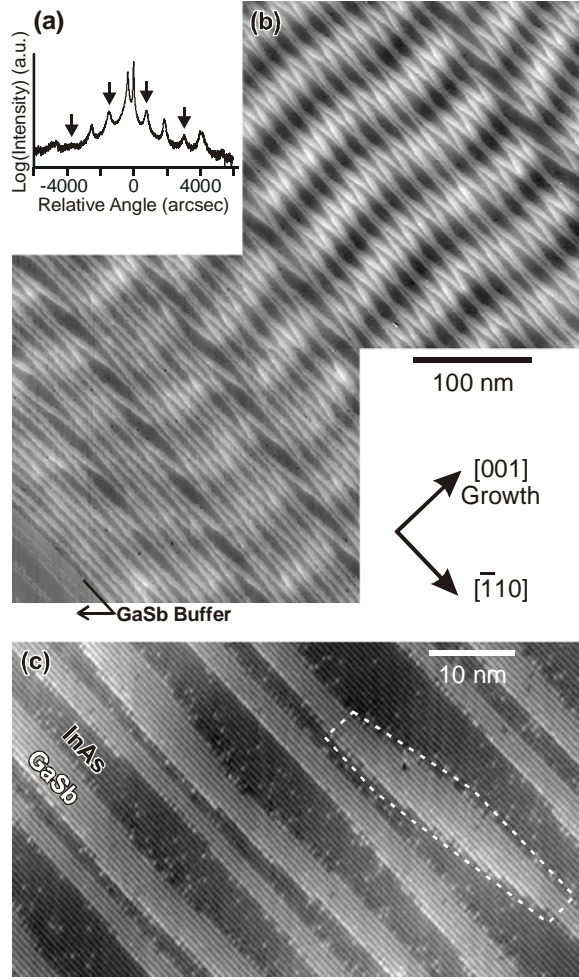


FIG. 1. A vertically-organized, lateral composition modulation observed in an InAs/GaSb superlattice grown with an $\text{As}_4\text{:In}$ flux ratio of 2.7:1. (a) X-ray diffraction spectrum plotted relative to the Bragg angle for bulk GaSb. The “extra” satellite peaks corresponding to twice the intended period are indicated. (b) XSTM image of the (110) cleavage plane through the first 80 periods of the superlattice. (c) Atomic resolution image (filled states at 2.1 V, 120 pA). The outlined area highlights the thickening of the GaSb along the stepped edge of the InAs mound that is characteristic of a vertically well-ordered structure.

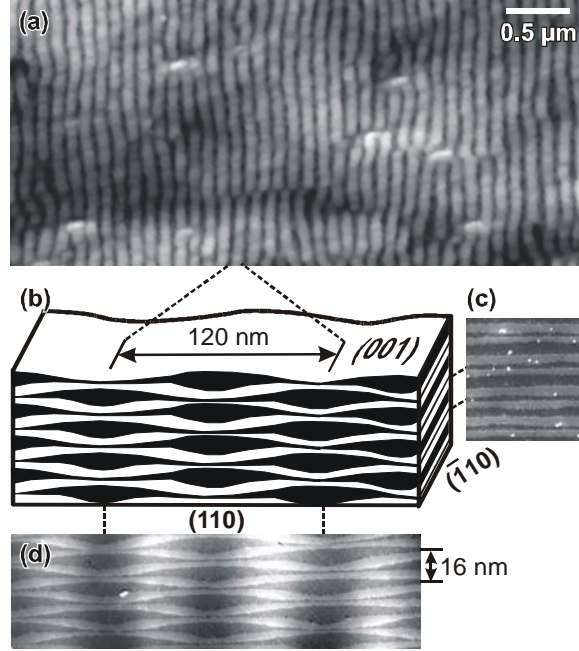


FIG. 2. The three-dimensional structure of the InAs nanowire lattice. (a) Atomic force microscopy image of the growth surface. (b) Illustration of the structure (traced from actual lateral modulations). (c) XSTM image of a $(\bar{1}10)$ surface. (d) A (110) surface.

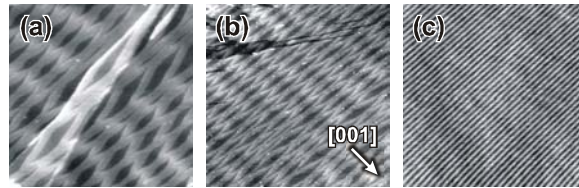


FIG. 3. XSTM images of (110) cross-sections through superlattices grown with the same As_4 flux but different periods or buffer layers, as follows: (a) doubled GaSb layers (26 ML thick); (b) relaxed AlSb buffer; (c) InAs buffer and substrate.